A-priori analysis of the flame structure in thermodiffusively unstable NH3/H2/air mixtures

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Abstract
In this investigation, we adopt a data-driven approach utilizing Direct Numerical Simulation (DNS) data from thermodiffusively unstable NH3/H2-Air mixtures to define progress variables, thereby addressing flame dimensionality concerns. Irreducible error analysis highlights the necessity of incorporating at least three progress variables for a comprehensive depiction of hydrogen-enriched ammonia flames. While two-dimensional models suffice for accurately representing temperature, three-dimensional models are essential for effectively reproducing NO dynamics. Additionally, our investigation of Artificial Neural Network (ANN) models reveals their ability to faithfully replicate DNS data when trained on a subset of that data. However, their performance markedly declines when exclusively trained on a dataset comprising unstretched 1D freely propagating flames. This suggests the critical importance of expanding datasets, particularly by incorporating stretched flames, to bolster the ANN model's capacity.

Introduction
Ammonia (NH3) is a promising energy carrier due to its high volumetric concentration of hydrogen (H2), making it efficient for energy storage and transportation. Its clean-burning nature with zero-carbon emissions has garnered attention as a potential solution to environmental concerns linked with fossil fuels [1]. However, challenges exist for its direct utilization as a fuel, notably its low reactivity and the production of nitrogen oxides (NOx) during combustion, particularly in stationary gas turbines. Strategies like blending with hydrogen or partial cracking aim to enhance reactivity [2], while adopting lean premixed combustion can minimize pollutant emissions.
The efficacy of these strategies depends on factors such as fuel mixture composition and combustion conditions. For instance, the effective Lewis number Le_{eff} of NH3/H2 blends is critical, influencing flame stability and combustion characteristics, particularly in lean mixtures where Le_{eff} falls below a critical threshold Le_0 [3]. Under such conditions, intrinsic flame instabilities (IFI) may occur, affecting various flame features, heat release rates, morphology, propagation, and pollutant formation [4]. These instabilities are governed by two mechanisms: the destabilizing hydrodynamic (Darrieus-Landau, DL) mechanism and the

stabilizing/destabilizing thermal-diffusive (TD) mechanism, driven by local imbalances between thermal conductivity and molecular diffusivity of the controlling reactant [5]. Addressing the flame dimensionality issue is crucial for understanding and modeling these complex combustion processes. One method involves using tabulated chemistry, assuming all relevant chemical properties lie on a low-dimensional manifold (LDM) described by a limited number of transported scalars $\Psi = \psi(C_1, C_2, \ldots, C_n)$. It has been shown that at least two-dimensional LDM is needed to accurately reproduce the non linearities and the fluctuations generated form the IFIs in pure hydrogen premixed flames [6], and many investigations have been carried out that proposed different parametrization. Lapenna et al. [7] proposed a representation in terms of non-dimensional temperature Θ and mass fraction of the deficient reactant Y_{H2} using a simplified irreversible one-step chemistry formulation, while Regele et al. [8] proposed a LDM based on the normalized water vapor mass fraction $Y_{H2O}$and a mixture fraction Z.

In this study, we employ a data-driven methodology using Direct Numerical Simulation (DNS) data from thermodiffusively unstable NH3/H2-Air mixtures. Our goal is to identify relevant "data-driven" Progress Variables by applying Singular Value Decomposition (SVD) to matrices containing chemical species data from DNS or 1D datasets generated using Cantera [9]. Through SVD, we derive Progress Variables that capture correlations within the chemical composition space. These variables are used to reproduce target observables from a DNS dataset, with differences analyzed among various reproduction methodologies. Specifically, Progress Variable definitions extracted via SVD from the training dataset are applied to the testing DNS dataset. Our results show that a model directly trained on DNS data outperforms one trained on a 1D dataset in replicating the DNS dataset. Additionally, through irreducible error analysis, we demonstrate that a minimum of three Progress Variables are needed for a comprehensive description of a hydrogen-enriched ammonia flame and for accurately reconstructing its structural characteristics.

Data and Methods

To evaluate the Low-Dimensional Manifold (LDM) dimension for ammonia/hydrogen mixtures, we utilize two distinct datasets. Firstly, we employ the AH1s DNS developed by D'Alessio et al. [10]. This dataset comprises statistically planar premixed flames consisting of 50% NH3 and 50% H2 by volume in air, operating at an equivalence ratio of $\Phi = 0.5$. These two-dimensional flames are simulated within rectangular computational domains that exhibit periodicity in the transverse direction. A small subset of the DNS is used for training while the whole DNS dataset serves as the final target. Secondly, we utilize a dataset generated via Cantera, comprising 30 unstretched freely propagating one-dimensional flame solutions. These solutions were obtained by spanning the equivalence ratios values in the range $\Phi = [0.45, 0.55]$ in order to capture the local variations of equivalence ratio experienced by an unstable 2D flame due to the presence of the instabilities.

Following the methodology outlined in Najafi et al. (2012) [11], singular value decomposition (SVD) is employed to systematically identify a subset of progress variables that effectively capture the Direct Numerical Simulation (DNS) data while minimizing variance. The DNS data, represented as species mass fractions, is structured into a data matrix denoted as X, upon which SVD is performed, resulting in the decomposition $X = U\Sigma V^T$. Progress variables are subsequently derived from the unitary matrix V^T. Up to four progress variables are denoted as $C_j = \Sigma_i a_{i,j} Y_i$, where $a_{i,j}$ is the weight of species i in progress variable j.

Following the determination of Progress Variables, the concept of the optimal estimator, as elucidated in Trisjono et al. (2015) [12], is applied utilizing the two quantities of interest (QoI) being the temperature T and the NO mass fraction Y_NO. This estimator may adopt diverse forms, such as the conditional average of the QoI concerning the set of progress variables $C_{1,\dots,q}$. For this investigation, aligned with Berger et al. (2018) [6], an artificial neural network (ANN) is trained utilizing 10,000 randomly chosen samples from a DNS field or a 1D flames dataset. The ANN architecture comprises three hidden layers housing 5 neurons each, utilizing sigmoid as activation functions, while the output layer employs a linear activation function. These architectural parameters were judiciously selected to ensure the convergence of results. In particular, the sampling procedure was aided by means of the Kullback-Liebler divergence, employed to evaluate the difference between the dataset and the sample's probability distribution functions. This approach allowed us to select a reasonable sample dimension that was a good compromise between ease of training and retained information. The resultant ANN serves as a surrogate for the optimal estimator of the QoIs, denoted as $\zeta^{ANN,q}$, where q denotes the total count of progress variables employed to represent the QoI.

Subsequently, the irreducible error associated with the optimal estimator is computed by contrasting it with the DNS field, yielding:

$$\epsilon_{irr} = < (\zeta^{TRUE} - \zeta^{ANN,q})^2 | C_{1,\dots,q} > \quad (1)$$

The irreducible error is standardized by the maximum value of the conditionally averaged $\omega_{\dot{N}O}$ with respect to the primary scalar C_1, expressed as:

$$\epsilon_{irr,norm} = \frac{\sqrt{\epsilon_{irr}}}{max(<\omega_{\dot{N}O}|C_1>)} \quad (2)$$

Results and discussion

The analysis of the Irreducible Error for each QoI shows decreasing values as the model dimensionality increases, allowing the assessment of the number of needed Progress Variables to achieve a desired accuracy in the reconstruction. For the QoI Temperature, the Irreducible Error significantly decreases while passing from mono-dimensional models to two-dimensional models, but stops significantly decreasing if models with three or more variables are used. This indicates that two dimensional

models are more than sufficient to achieve an excellent representation of Temperature. For the variable NO the Irreducible Error analysis suggests that at least three variables are required to obtain good reconstruction results as shown in Tab1.

Table 1. Average Irreducible Error normalized by maximum QoI value, according to dimensionality of the model.

	1D	2D	3D	4D
Temperature	0.0526	0.0080	0.0078	0.0042
Y_NO	0.1259	0.0397	0.0149	0.0141

The Artificial Neural Network (ANN) model was initially trained on the DNS dataset to effectively reproduce the specified Quantities of Interest (QoIs). The outcomes, depicted in Fig.1, demonstrate remarkable fidelity, particularly when employing two or more Progress Variables for temperature, and three or more for NO. Subsequently, the same model trained on the 1D dataset was applied to the DNS testing dataset. As illustrated in Fig.2, while the model exhibits comparable results for temperature, it consistently falls short in accurately reproducing NO. Notably, augmenting the dimensionality does not seem to enhance the model's capacity for NO reproduction. This observation prompts the inference that an expanded 1D dataset, incorporating stretched flames, is indispensable for attaining improved results, particularly concerning NO reproduction.

Conclusion

In conclusion, our study utilized a data-driven model combining SVD-derived Progress Variables and ANN interpolation. Trained on both DNS and 1D datasets, the model aimed to replicate NO mass fraction and temperature from a reference DNS dataset. Irreducible Error analysis revealed that at least two Progress Variables were necessary for temperature modeling, and three for NO.

Temperature dynamics were accurately replicated by models trained on both DNS and 1D datasets with at least two Progress Variables. However, superior NO reproduction was achieved only by models directly trained on DNS data, requiring three Progress Variables. Conversely, models trained solely on 1D data showed suboptimal performance in capturing NO behavior.

While the model trained on freely propagating flames proficiently reproduced temperature dynamics, it struggled to accurately characterize NO behavior in unstable flames. Future studies should explore incorporating stretched 1D flames to enhance NO reproduction capability. Overall, our findings contribute to advancing flame modeling understanding, emphasizing the importance of addressing complexities highlighted by Irreducible Error analysis.

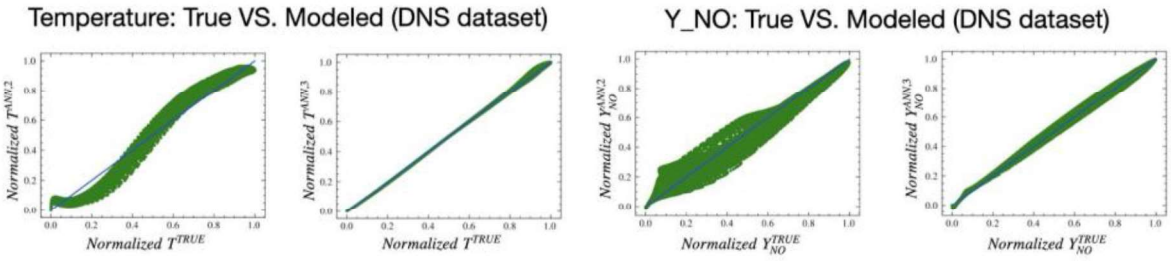

Figure 1. Apriori scatters of presented QoIs. Each plot presents x-axis as the scaled DNS reference dataset values, and the y axis as the values retrieved by the model trained onto the DNS dataset. Above is NO data for 2D and 3D models, while below is Temperature data for 1D and 2D models.

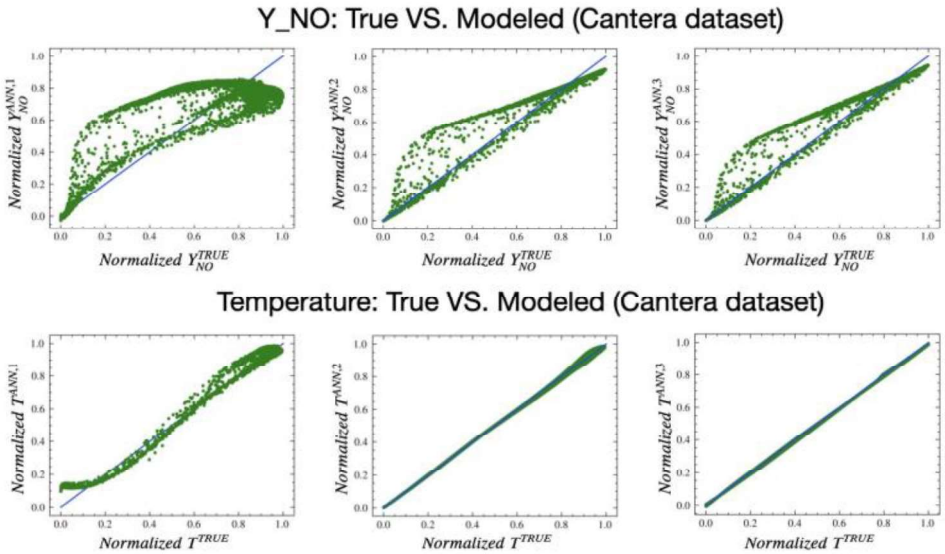

Figure2. Apriori scatters of presented QoIs. Each plot presents x-axis as the scaled DNS reference dataset values, and the y axis as the values retrieved by the model trained onto the 1D dataset. Above is NO data for 1D, 2D and 3D models, while below is Temperature data for 1D and 2D models.

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